

RESEARCH ARTICLE

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The use of the GIS Kriging technique to determine the spatial changes of natural radionuclide concentrations in soil and forest cover

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Abstract

Background: The distribution of radionuclides occurring naturally in the earth depends on bedrock characteristics. Therefore, the spatial distribution of radionuclides is not uniform. Consequently, radionuclide information is vitally important in determining and monitoring the spatial variation of the radionuclide concentrations that are over the limits for the sustainable environment and human health.

Methods: This research was carried out using GIS methods and geostatistical analysis as Kriging techniques to reveal the spatial variation of the ^{226}Ra , ^{232}Th and ^{40}K concentrations of natural radionuclides in the Çoruh and Aras Basin. The spatial variations of the detected radionuclides were correlated with soil groups and forest cover.

Results: In the study area, 43.17% of the concentration of ^{40}K , 26.67% of the concentration of ^{226}Ra and 28.16% of the ^{232}Th concentration was determined to be over the average limits. Concentrations of radionuclides that are over the average limits have been found to be on basalt and chestnut soils. Brown and reddish brown soils have a low concentration of the spatial distribution of the radionuclides. Statistically positive correlations were detected (0.865 **) between the ^{226}Ra and ^{232}Th . In addition, a positive relationship between forest cover and ^{226}Ra and a negative relationship between ^{232}Th and ^{40}K were identified.

Conclusions: Excessive exposure to radiation may cause cancer and hereditary diseases. Ecological environments include the soil and the plants. Hence, the periodical monitoring of the spatial variation in concentrations of radionuclides is very important for the health of future generations.

Keywords: Natural radionuclide, Kriging technique, Soil, Forest cover

Background

Radioactive features have existed in our world since its formation. High concentrations of natural radionuclides are found in volcanic, phosphate, granite and salt rock. Rain and other water discharge crumble these rocks into very small pieces and mixes them into the groundwater. Thus, rocks increase the natural radioactivity of the soil.

The direction of the movement and the speed of the radionuclides in the soil depends on natural processes (e.g., soil structure, content of the plant species, irrigation conditions, weather conditions and accumulation) [1,2]. In some areas, the natural radionuclide concentrations are above the established limits according to

UNSCEAR. If the concentration of natural radioactive elements goes over the average limits, there can be negative effects on human health [3]. Therefore, the spatial distribution of the concentrations of natural radionuclides in the soil has to be determined. It is possible to use CBS and geostatistical analysis to obtain these values.

Geostatistical techniques are a useful component of GIS applications that are frequently applied. Geostatistics involves the analysis and estimation techniques used to obtain the value of a variable dispersed in time and location.

Kriging is one of the best and most widely-known techniques used in spatial linear predictions. Kriging methods have different flexible forms, according to the survey area and data [4-7]. Kriging can also reveal the reliability of the estimated surface [8,9].

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Geostatistical methods also allow for examining a relationship with spatial variations of radionuclides between forest cover and soil groups. Radioactive elements in the soil do not indicate a uniform distribution in the earth. Therefore, the concentrations of radionuclides should be checked regularly as an important step in protection from the negative effects of radioactivity [10-12].

Radionuclide concentrations may be caused by the high amount of organic matter in the soil. As such, radionuclides can be absorbed by the forest soil [13,14]. Some radionuclide compounds build up in the humic acids in the soil organic layer [15,16].

Measuring the radioactivity concentration in the soil, as well as the concentrations in the plants and the water, is necessary to estimate the concentrations of radionuclides [17,18]. This study used ^{226}Ra , ^{232}Th and ^{40}K concentration values measured periodically by TAEK [19]. Spatial distributions were determined according to UNSCEAR (2000) [3], who used a kriging technique in the Çoruh and Aras Basin and the surrounding areas. The statistical relationships between the spatial variations, forest cover and soil groups were analyzed.

Materials and methods

This research was carried out in 10 provinces in eastern Turkey: Rize, Bayburt, Erzurum, Artvin, Ardahan, Kars, Iğdır, Muş, Bingöl and Erzincan. The bounding geographical coordinates of the study area are $39^{\circ}50'07''$ to $39^{\circ}46'17''$ north latitudes and $38^{\circ}26'52''$ to $44^{\circ}35'46''$ east longitudes (Figure 1). The study area is 9.815.000 ha in size. Forest areas were detected using local Forest Management Plan data [20].

Sampling methods and analysis

Concentrations of radioactive elements (^{226}Ra , ^{232}Th and ^{40}K) for determining soil sampling were carried out by the provincial offices of the Ministry of Environment and Urbanization. Surface soil sampling was conducted at 46 points. The concentration of radioactivity in the surface soil samples was determined by the Atomic Research Council of Turkey [19].

Risk assessment

The average natural radioactivity rate, the concentration range and the average values are presented in Table 1 [3]. To determine the concentration of the natural radionuclides, soil samples from 10 cm thickness in 1 km^2 land were collected in the research area.

Geostatistical analysis

To conduct the geostatistical analysis, the "Kriging" interpolation technique was used within the spatial analyst extension module in ArcGIS 9.3 software. The spatial analyses were carried out with prepared maps

using this technique. Concentrations of ^{226}Ra , ^{232}Th and ^{40}K were determined for the distribution area. The experimental variogram model was constructed using the Kriging method, with data obtained from the research area. The spatial transformation was performed to determine the most appropriate model to use with the parameters of the generated maps.

The ordinary Kriging formula is as follows [8,21]:

$$Z(S_0) = \sum_{i=1}^N \lambda_i Z(S_i)$$

where:

$Z(s_i)$ is the measured value at the location (i th),

λ_i is the unknown weight for the measured value at the location (i th) and

s_0 is the estimation location.

The unknown weight (λ_p) depends on the distance to the location of the prediction and the spatial relationships among the measured values.

The statistical model estimates the unmeasured values using known values. A small difference occurs between the true value $Z(s_0)$ and the predicted value, $\sum \lambda_i Z(s_{ij})$. Therefore, the statistical prediction is minimized using the following formula:

$$\left[Z(S_0) - \sum_{i=1}^N \lambda_i Z(S_i) \right]^2$$

The Kriging interpolation technique is made possible by transferring data into the GIS environment. In this way, analysis in areas that have no data can be conducted. The following criteria were used to evaluate the model: the average error (ME) must be close to 0 and the square root of the estimated error of the mean standardized (RMSS) must be close to 1 [22]. While implementing the models, the anisotropy effect was surveyed.

Results and discussion

Anisotropic variogram models were preferred. The ^{226}Ra , ^{232}Th and ^{40}K concentration values showed a directional change. The spatial dependencies (Nugget/Sill ratio) were found to be related to the degree of autocorrelation between the sampling points. If the spatial dependence was higher between the sample points, the spatial correlation was very high. The spatially dependent variables were classified as: strongly spatially dependent if the ratio was $\leq 25\%$, mid-spatial-dependent if the ratio was $25\% - 75\%$ and weakly spatially dependent if the ratio was $\geq 75\%$ [4,23-26].

Because the spatial dependence was strong, the variables did not differ over short distances. In this research area, spatial dependence was too high for ^{232}Th (16.91%).

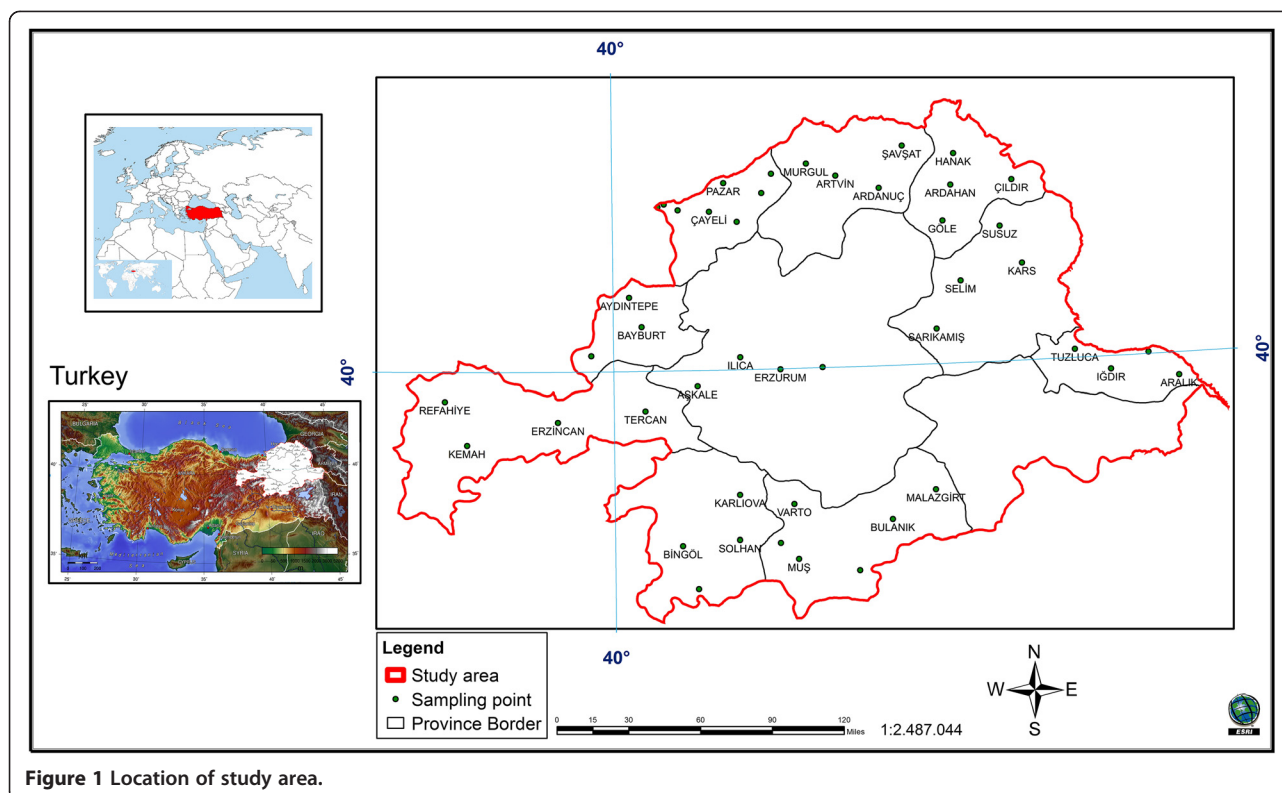


Figure 1 Location of study area.

Spatial dependence was identified as normal for ^{226}Ra and ^{40}K (25.88% and 70.60%), respectively. An effective spatial dependence distance was found between 254369.7 and 375276 meters (Table 2).

The sample point data will involve converting the spatial data of the ^{226}Ra , ^{232}Th and ^{40}K concentrations used in the interpolation for kriging. The lowest error rate models were chosen; they were the “Exponential” and “Stable” models. The maps were produced and field data were obtained in accordance with this kriging model.

Spatial distribution of ^{226}Ra :

The prediction map, according to the optimized model, was determined during the cross-validation process. The ^{226}Ra concentration prediction map shows the $\log^{226}\text{Ra}$. The dataset for the ^{226}Ra concentration has a high kurtosis and is positively skewed, so it is not a normal distribution. The data log transformation was applied

to be closer to a normal distribution. After the log transformation was conducted, the ^{226}Ra concentrations were found to be approximately normally distributed.

The histogram of ^{226}Ra and log transformations data is presented in Figure 2. To check the data, the best model was applied to the cross-validation of the spatial correlation of the ^{226}Ra concentration of the study area. A comparison of the ME and the RMSE for the $\log^{226}\text{Ra}$ illustrates that the exponential model and its parameters were best for the ^{226}Ra concentration. The exponential model has the best fit with the nugget effect (C_0); it is equal to 48.35, a sill ($C_0 + C$) equal to 186.83 and a range of influence equal to 254369.7. The ratio of the nugget variance to the sill is expressed in percentages equal to 25.88% (Table 2). This value is greater than 25% and less than 75%. Thus, the ^{226}Ra distribution has a moderate spatial dependence in the study area. A spatial prediction and distribution map for the ordinary kriging interpolation of ^{226}Ra is presented in Figure 3.

Table 1 Natural radioactivity the concentration range and average values

| | Natural radionuclides (Bq/kg) | | |
|---------------------|-------------------------------|-------------------|-----------------|
| | ^{226}Ra | ^{232}Th | ^{40}K |
| Concentration range | 17-60 | 11-64 | 140-850 |
| Average values | 35 | 45 | 400 |

Spatial distribution of ^{232}Th

The dataset of the ^{232}Th concentration has a high kurtosis and is positively skewed, so it is not a normal distribution. The data log transformation was applied, so could be closer to a normal distribution. After the log transformation was conducted, the ^{232}Th concentrations were approximately normally distributed. The histogram

Table 2 Model parameters

| | Model | Regression function | Nugget, (C _o) | Range, A | Sill, (C _o + C) | Nugget/Sill, (%) | ME ¹ | RMSSE ² |
|-------------------|-------------|---------------------|---------------------------|----------|----------------------------|------------------|-----------------|--------------------|
| ²²⁶ Ra | Exponential | 0.2695 x +18.32 | 48.35 | 254369.7 | 186.83 | 25.88 | 0.004 | 1.06 |
| ²³² Th | Exponential | 0.5063 x +15.33 | 50.74 | 304696.1 | 300.03 | 16.91 | 0.008 | 0.89 |
| ⁴⁰ K | Stable | 0.1596 x +381.99 | 41767.52 | 375276.0 | 59158.77 | 70.60 | 0.007 | 0.997 |

ME¹: mean standard error.

RMSSE²: estimated standardized mean of error of mean square root.

of ²³²Th and log-histogram transformation data is presented in Figure 4. To check that the best model was applied, a cross-validation of the spatial correlation of the ²³²Th concentration of the study area was conducted. A comparison of ME and the RMSSE for log ²³²Th shows that the exponential model and its parameters are the more appropriate for the ²³²Th concentration. The exponential model has the best fit, with the nugget effect (C_o) being equal to 50.74, a sill (C_o + C) equal to 300.03 and a range of influence equal to 304696.1. The ratio of the nugget variance to the sill expressed in percentages is equal to 16.91% (Table 2). This value is smaller than 25; thus, the ²³²Th distribution has a powerful spatial dependence in the study area. A spatial prediction and

distribution map for the ordinary kriging interpolation ²³²Th is presented in Figure 5.

Spatial distribution of ⁴⁰K

An optimized model determined from the cross-validation process and the ⁴⁰K concentration prediction map shows the log ⁴⁰K. The dataset for the ⁴⁰K concentration has a high kurtosis and is positively skewed, so it is not a normal distribution. The data log transformation was applied, so the data would be closer to a normal distribution. After the log transformation, the ⁴⁰K concentrations were found to be approximately normally distributed. The histogram of ⁴⁰K and the log transformation data is presented in Figure 6.

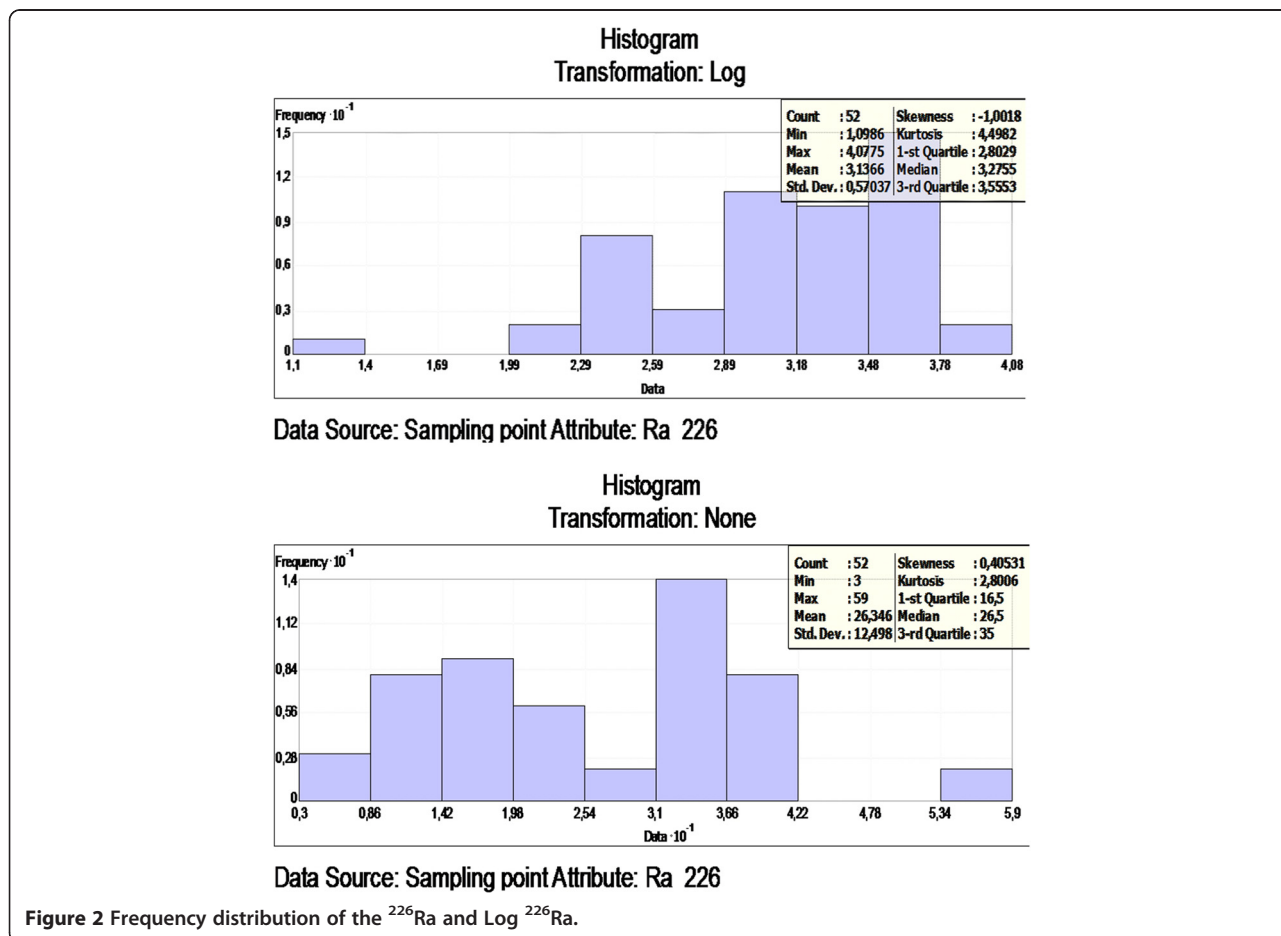
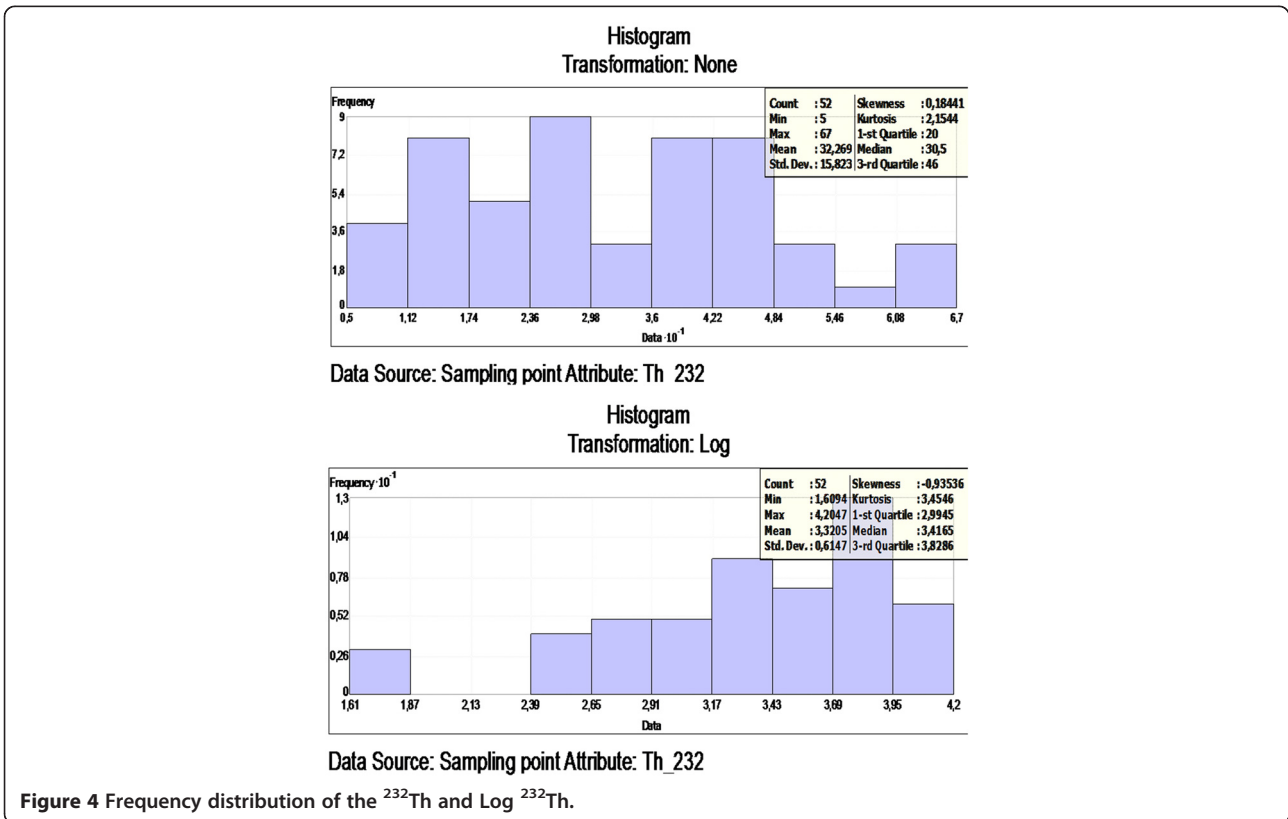
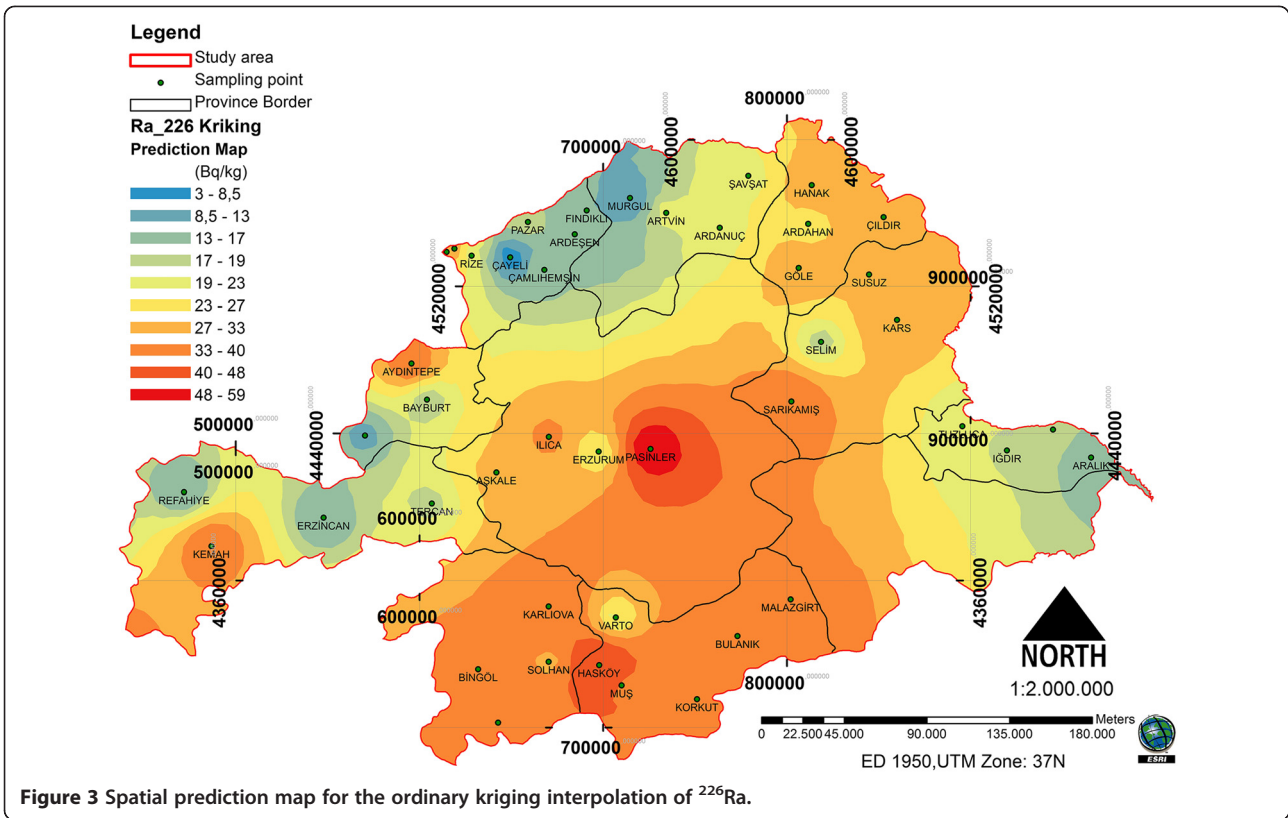
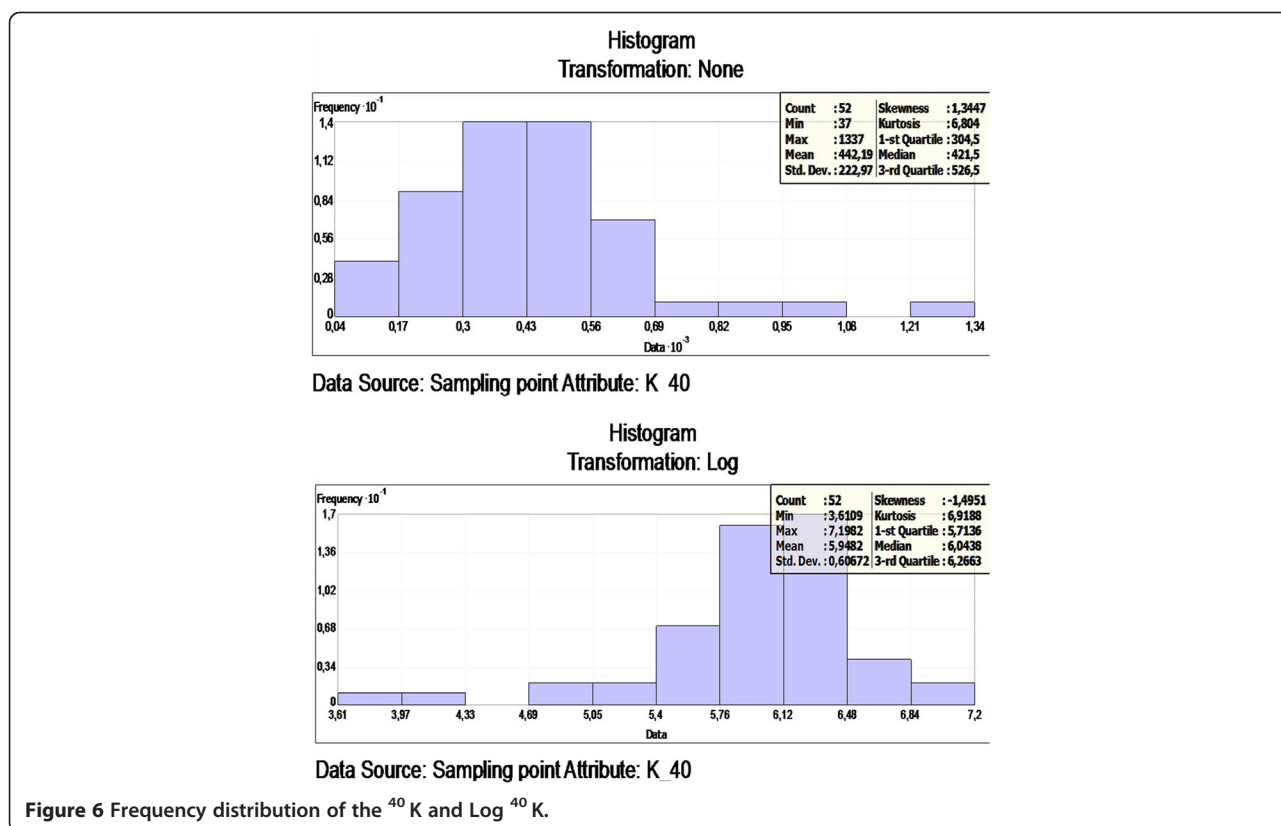
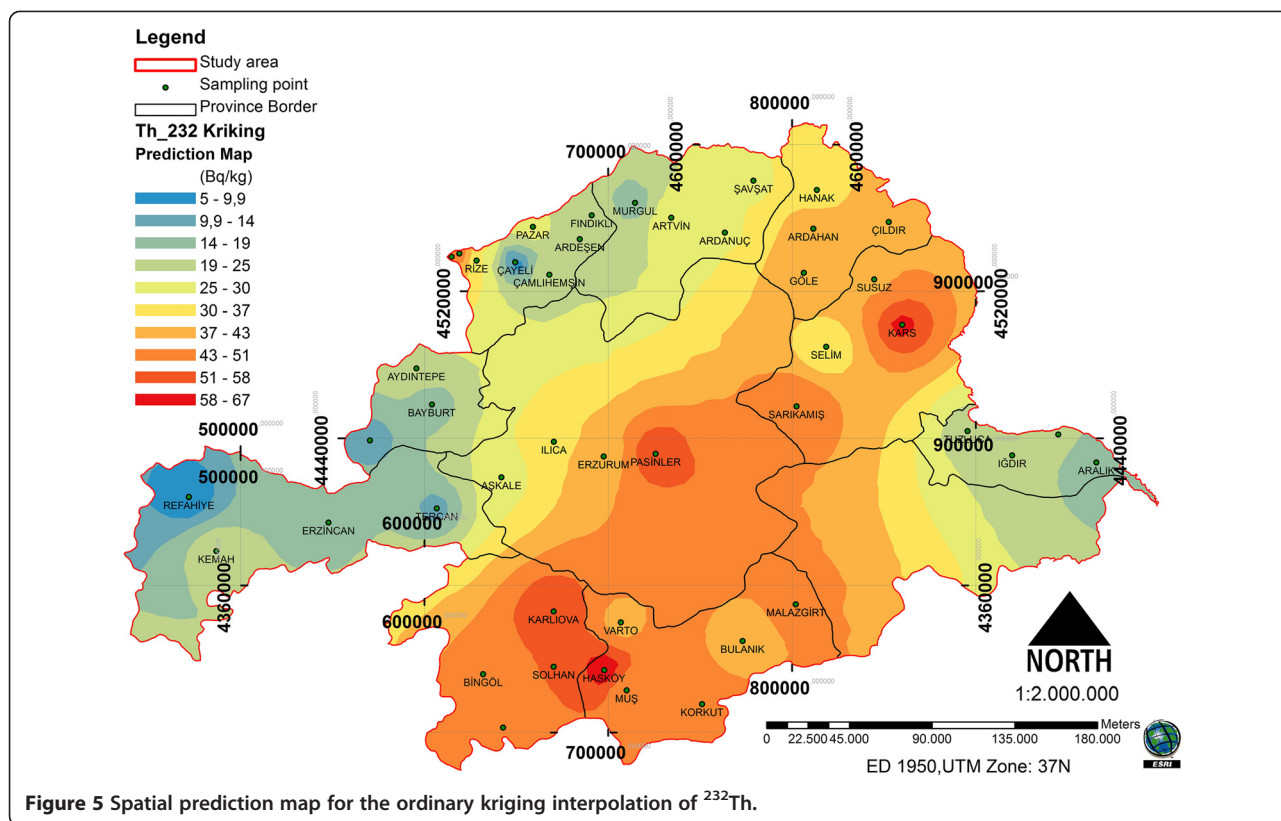
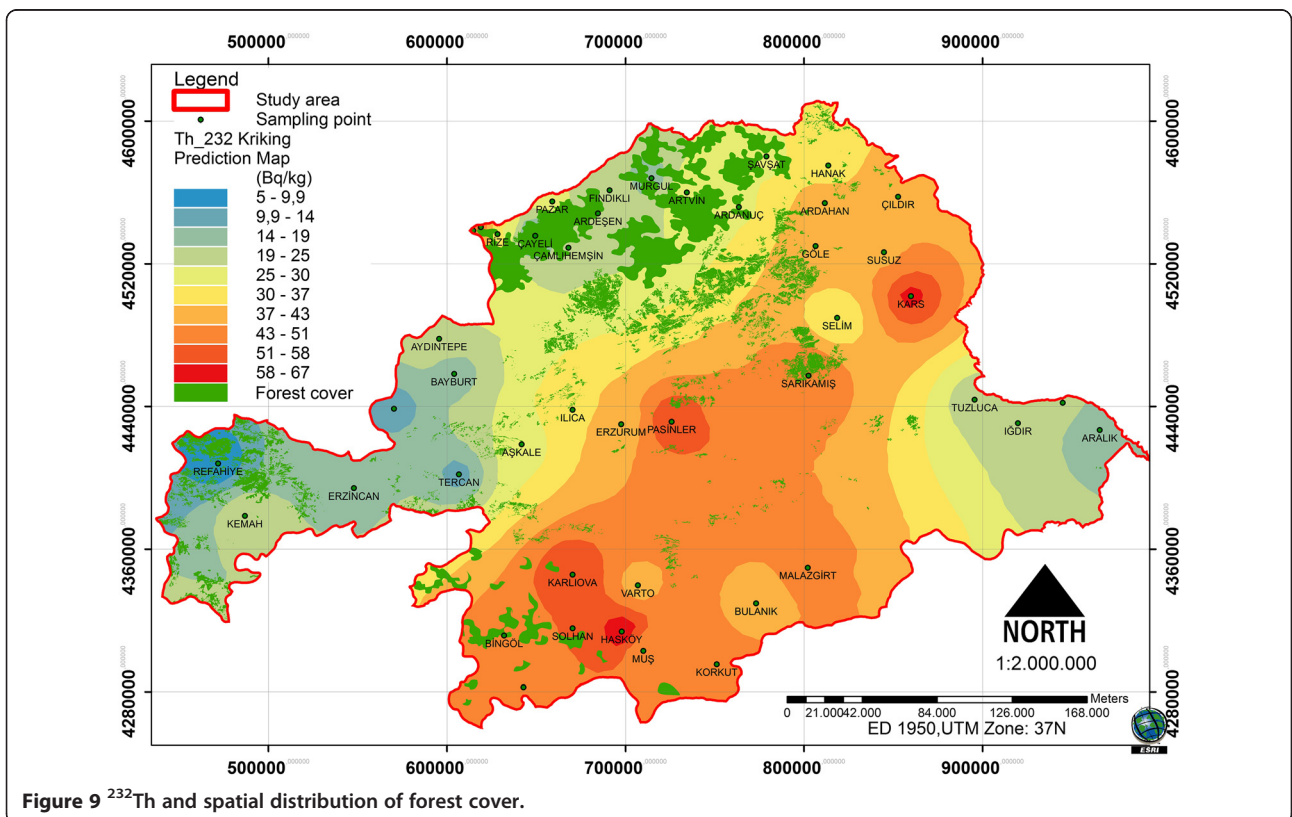
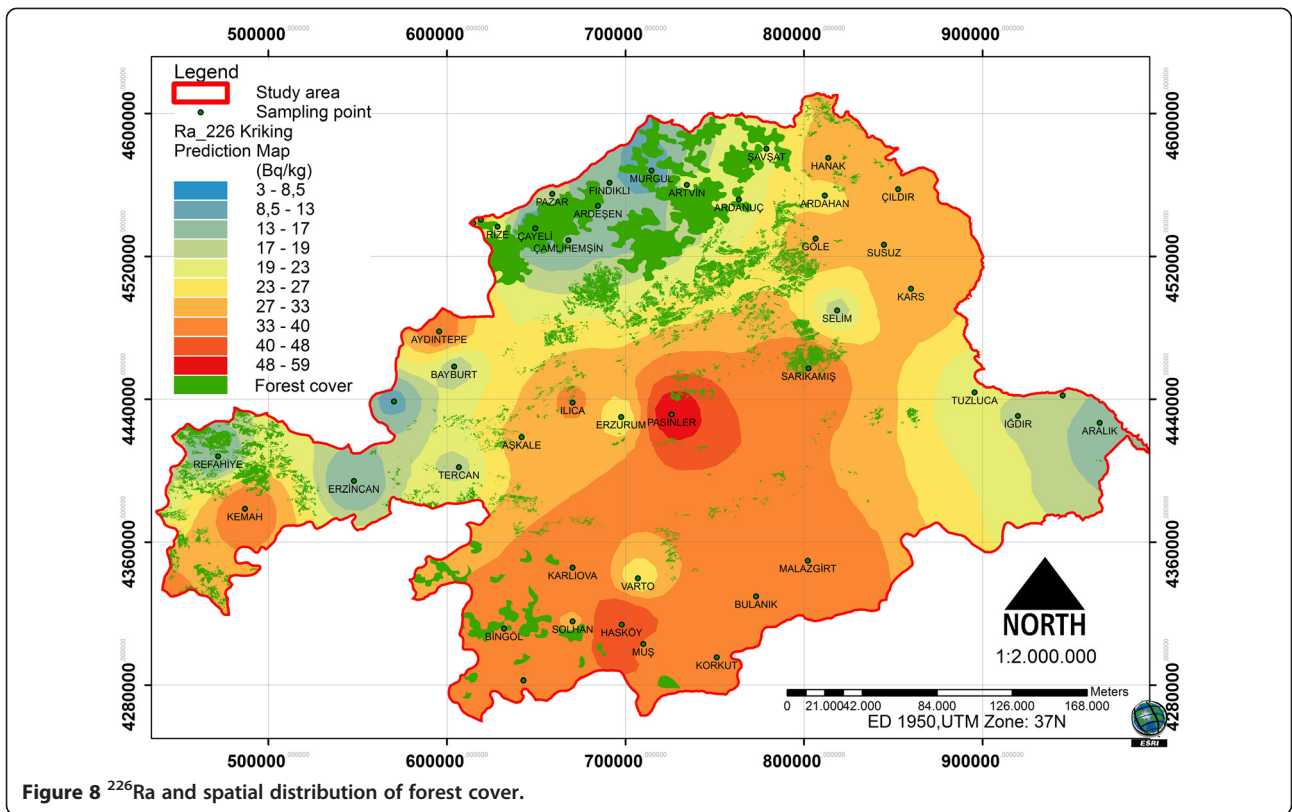
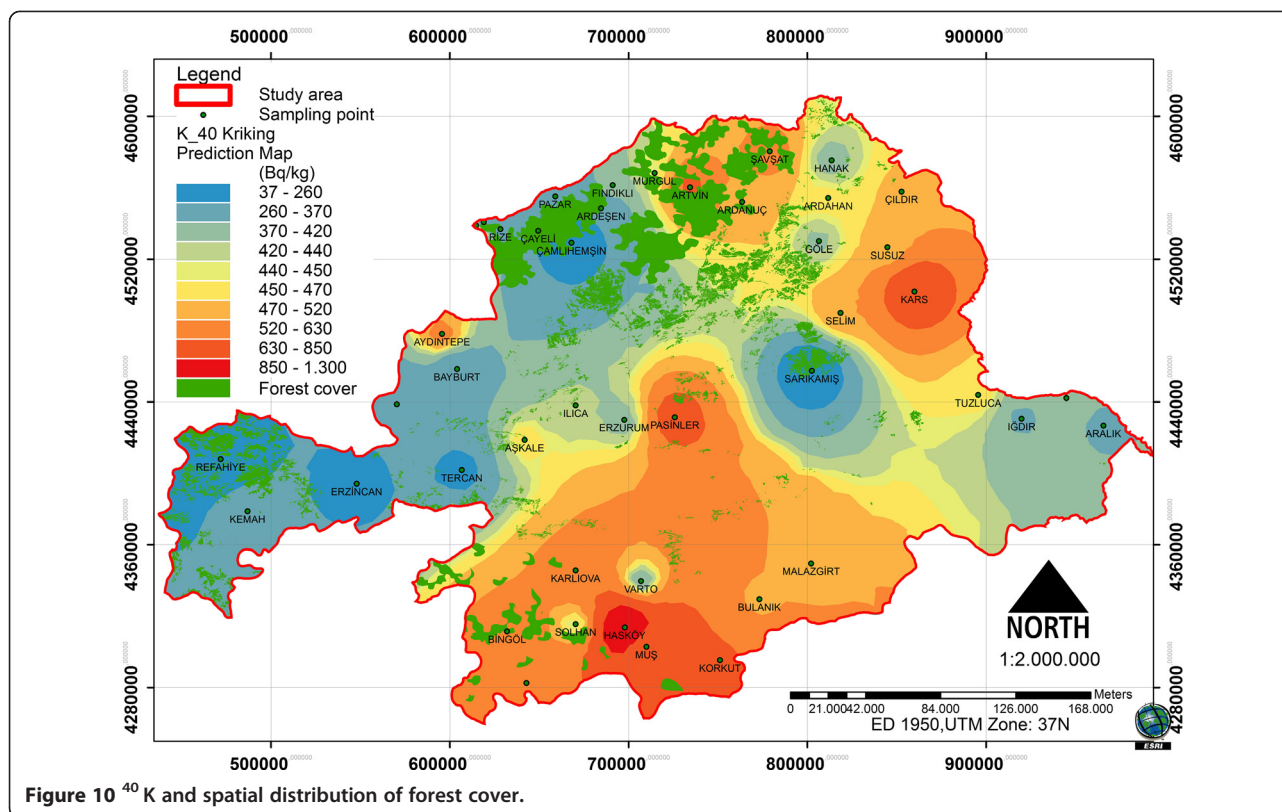


Figure 2 Frequency distribution of the ²²⁶Ra and Log ²²⁶Ra.









In summary, changes in the concentrations of radionuclides in the soil depend on the formation of iron oxide and other elements. Some of the acids formed in the soil, through calcium carbonate found in the environment, prevent the retention of the radionuclides. Therefore, radionuclide concentrations in the rocks can be reduced with calcium carbonate; this, in turn, reduces the level of external radiation [28,29]. According to the Anonymous [30], some rocks and a soil type typical of the specific radioactivity values was identified in the follow: For ⁴⁰K; Granite (1000 Bq/kg), clay stone (700 Bq/kg), Sandstone (350 Bq/kg) Basalt (250 Bq/kg) and limestone (90 Bq/kg). For ²³²Th; Granite (80 Bq/kg), clay stone (50 Bq/kg), Sandstone (10 Bq/kg), Basalt (10 Bq/kg) and Limestone (7 Bq/kg). Local

distribution values can vary greatly according to changing areas [31,32].

Conclusions

Over time, the infiltration of radionuclides has resulted in high radionuclide concentrations in the lower soil layers. The radionuclides in these lower soil layers can move upwards through the roots of plants and be transferred to the plant during the growth process. Since radionuclides can have detrimental health effects on humans, it is important to determine the spatial variation of concentrations of radioactive elements.

This research was conducted to examine to spatial distribution of natural radioactive element (²²⁶Ra, ²³²Th and ⁴⁰K) concentrations and their relationship with soil groups and forest cover using the Kriging method. According to the statistical analyses, positive correlations were detected between the ²²⁶Ra and the ²³²Th (0.865**), as well as between the ⁴⁰K and ²³²Th (0.718*). Negative correlations between forest cover and ²²⁶Ra were found, while positive correlations between ²³²Th and ⁴⁰K were detected.

The basalt and chestnut soils in the study area were found to have above average concentrations of radionuclides. The Brown soils, High Mountain soils, Reddish Brown soils and Reddish Yellow Podsol soils

Table 3 Correlations test result between radionuclides and forest cover

| Parameters | Radionuclides | | | Forest cover |
|-------------------|-------------------|-------------------|-----------------|--------------|
| | ²²⁶ Ra | ²³² Th | ⁴⁰ K | |
| ²²⁶ Ra | 1 | | | |
| ²³² Th | 0,865** | 1 | | |
| ⁴⁰ K | 0,592 | 0,718* | 1 | |
| Forest cover | -0,008 | 0,096 | 0,232 | 1 |

**Correlation is significant at the 0.01 level (2-tailed).

*Correlation is significant at the 0.05 level (2-tailed).

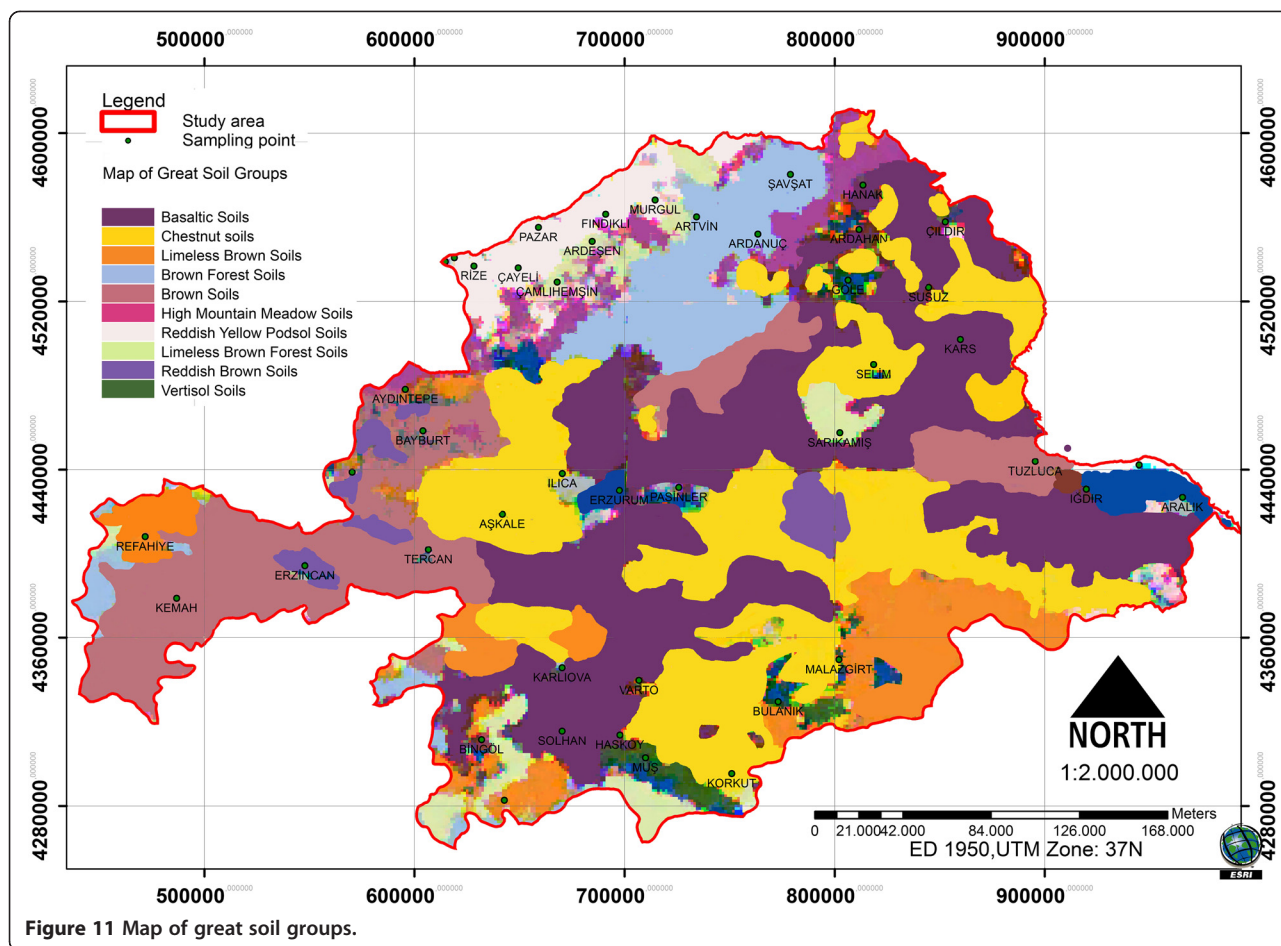


Figure 11 Map of great soil groups.

did not have high concentrations of ^{226}Ra and ^{232}Th . The Brown soils and the Reddish Brown soils also did not have high concentrations of ^{40}K .

Radionuclides are present in different concentrations in the soil, plants and water, which comprise parts of the

basic food chain. Excessive exposure to radiation can lead to cancer; it is also the cause of hereditary diseases. Therefore, spatial variations of radioactive element concentrations need to be monitoring for the sustainability of a healthy environment.

Table 4 Spatial changes between radionuclides and great soil groups

| Great soil groups | Spatial changes of the radionuclides concentration (Hectare) | | | | | |
|-----------------------------|--|---|--|--|--|--|
| | ^{40}K | | ^{226}Ra | | ^{232}Th | |
| | Under optimal concentrations (<400 Bg/kg) | Upper optimal concentrations (>400 Bg/kg) | Under optimal concentrations (<35 Bg/kg) | Upper optimal concentrations (>35 Bg/kg) | Under optimal concentrations (<45 Bg/kg) | Upper optimal concentrations (>45 Bg/kg) |
| Basaltic Soils | 1187656 | 1717242 | 1915127 | 989771 | 1876826 | 1028072 |
| Chesnut Soils | 1101871 | 1129844 | 1290559 | 941156 | 1316923 | 914792 |
| Limeless Brownish Soils | 428955 | 582383 | 620178 | 391160 | 762213 | 249125 |
| Limeless Brown Forest Soils | 94789 | 228346 | 119664 | 203471 | 135446 | 187689 |
| Vertisol Soils | 0 | 76254 | 0 | 76254 | 0 | 76254 |
| Brown Forest Soils | 464221 | 287303 | 684116 | 67408 | 684116 | 67408 |
| Brown Soils | 1505651 | 0 | 1505651 | 0 | 1505651 | 0 |
| High Mountain Soils | 262129 | 159430 | 421559 | 0 | 421559 | 0 |
| Reddish Yellow Podsol Soils | 438377 | 56680 | 495057 | 0 | 495057 | 0 |
| Reddish Brown Soils | 94684 | 0 | 0 | 94684 | 0 | 94684 |

Competing interests

The author declares that he has no competing interests.

Author's contribution

TD participated in the design of the study and has collected the data and drafted the manuscript used the GIS and makes statistical analysis related data with soil groups and forest area.

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